Available online at www.sciencedirect.com
ScienceDirect
Journal of Computational and Applied Mathematics 206 (2007) 534–548
JOURNAL OF COMPUTATIONAL AND APPLIED MATHEMATICS
www.elsevier.com/locate/cam

# An iterative langevin solution for turbulent dispersion in the atmosphere

Jonas C. Carvalho[a,*], Marco Túllio M.B. de Vilhena[b], Mark Thompson[b]

[a] Universidade Federal de Pelotas, Faculdade de Meteorologia, PPGMET, 96010-900, Pelotas, RS, Brazil
[b] Universidade Federal do Rio Grande do Sul, Instituto de Matemática, 91509-900, Porto Alegre, RS, Brazil

Received 13 February 2006; received in revised form 25 June 2006

## Abstract

In this work we present an alternative hybrid method to solve the Langevin equation and we apply it to simulate air pollution dispersion in inhomogeneous turbulence conditions. The method solves the Langevin equation, in semi-analytical manner, by the method of successive approximations or Picard's Iterative Method. Solutions for Gaussian and non-Gaussian turbulence conditions, considering Gaussian, bi-Gaussian and Gram–Charlier probability density functions are obtained. The models are applied to study the pollutant dispersion in all atmospheric stability and in low-wind speed condition. The proposed approach is evaluated through the comparison with experimental data and results from other different dispersion models. A statistical analysis reveals that the model simulates very well the experimental data and presents results comparable or even better than ones obtained by the other models.


*Keywords:* Atmospheric dispersion; Langevin equation; Lagrangian particle model; Picard's iteration method; Probability density function; Model evaluation

## 1. Introduction

Turbulent dispersion of scalars in the planetary boundary layer (PBL) is probably best understood in a Lagrangian framework as first suggested by Taylor's statistical theory of dispersion in homogeneous turbulence [47]. Lagrangian particle models are an important and effective tool to simulate the atmospheric dispersion of airborne pollutants. These models are based on the Langevin equation, which is derived from the hypothesis that the velocity is given by the combination between a deterministic term and a stochastic term. The main advantages of these models in relation to the other techniques (similarity or gradient-transfer theory) are the simplicity, flexibility and the ability to incorporate temporal and spatial variations in turbulence properties [28,39]. As a consequence, important features of flow and turbulence fields (such as the vertical profiles of wind speed, standard deviation and higher order moments of wind speed fluctuation) can be included in the model, thus allowing a more accurate simulation without consuming excessive computer time.

The Langevin equation was the first example of a stochastic differential equation and it is normally integrated according to the rules of the Ito calculus [36]. Some special solutions of the Langevin equation are presented in [36,20].

* Corresponding author. Tel.: +555332776722; fax: +555332776722.
*E-mail address:* carvalho@pesquisador.cnpq.br (J.C. Carvalho).

0377-0427/$ - see front matter 
doi:10.1016/j.cam.2006.08.007

Rodean [36], for instance, describes the solution for stationary homogeneous turbulence as suggested in [27,26]. In this work we present an alternative hybrid method to solve the Langevin equation and we apply it to simulate air pollution dispersion in inhomogeneous turbulence conditions. The methodology of solution has been proposed in three recent papers in [14,12,13]. The technique solves the Langevin equation, in a semi-analytical manner, by the method of successive approximations or Picard's Iterative Method [7]. Solutions for Gaussian and non-Gaussian turbulence conditions, considering Gaussian, bi-Gaussian (linear combination of two Gaussians) and Gram–Charlier (expansion in series of Hermite polynomials) probability density functions (PDFs) are obtained. Using Gram–Charlier PDF, Gaussian and non-Gaussian turbulence conditions can be considered, according to Hermite polynomial order. By using this methodology, the pollutant dispersion in all atmospheric stabilities and during low-wind speed conditions are simulated. An important feature of this work relies on the mathematical analysis of existence and uniqueness of the solution which makes the work complete under mathematical point of view.

It is important to mention that using the Gaussian PDF, we come out with quadratic drift of velocity. The value for the initial value of velocity (velocity at $t = 0$) is determined considering the stochastic term as a source term of the first-order differential Langevin equation. Using the bi-Gaussian PDF, the procedure is similar, but now the initial value for the wind velocity is taken from the random value of a non-Gaussian distribution. For the Gram–Charlier PDF, the initial value for the wind velocity is a random value taken from a Gaussian distribution. When considering the case of low-wind speed, where the Eulerian autocorrelation function suggested in [19] appears naturally in the solution, the initial value for the wind velocity is taken from a Gaussian distribution.

The performance of the proposed approach is evaluated considering the values of ground-level concentration measured during four tracer experiments, which are conducted in different atmospheric stabilities and windy/low-wind speed conditions. Furthermore, the results attained by this method are compared with the ones obtained by other dispersion models. Statistical indices are calculated to compare the simulated and observed concentrations. To reach this goal, the work is outlined as follows: in Section 2 we discuss the Langevin model, in Section 3 we present the mathematical background for the existence and uniqueness of the proposed solution, in Section 4 we report the derivation of the iterative scheme for the PDFs Gaussian, bi-Gaussian and Gram–Charlier, in Section 5 we display the solution of asymptotic Langevin equation (random displacement model), in Section 6 we show solution for low-wind speed conditions and in Section 7 we quote the results of the numerical simulations.

## 2. Lagrangian stochastic particle model

The Lagrangian stochastic particle models are based on Langevin equation for the random acceleration. The three dimensional Langevin equation for inhomogeneous turbulence is

$$\frac{\mathrm{d}u_i}{\mathrm{d}t} = a_i(x_i, u_i)\,\mathrm{d}t + b_i(x_i, u_i)\xi_i(t), \tag{1a}$$

where $x$ is the displacement, $u_i$ is the turbulent velocity, $a_i(x_i, u_i)\,\mathrm{d}t$ is the deterministic term, $b_i(x_i, u_i)\xi_i(t)$ is the stochastic term and $\xi_i$ is a normally distributed (average 0 and variance $\mathrm{d}t$) random increment. The displacement of each particle is given by

$$\mathrm{d}x_i = (U_i + u_i)\,\mathrm{d}t, \tag{1b}$$

where $U$ is the mean wind velocity.

The deterministic coefficient $a_i$ depends on the non-conditional PDF, $P(x_i, u_i)$, of the Eulerian turbulent velocity and must be determined from the Fokker–Planck equation for stationary conditions [45]:

$$\frac{\partial(u_i P)}{\partial x_i} = -\frac{\partial(a_i P)}{\partial u_i} + \frac{1}{2}\frac{\partial^2(b_i^2 P)}{\partial u_i^2}. \tag{2}$$

This equation can be split into two equations:

$$a_i P = \frac{\partial}{\partial u_i}\left(\frac{b_i^2}{2} P\right) + \phi_i(x_i, u_i) \tag{3a}$$

and

$$\frac{\partial \phi_i}{\partial u_i} = -\frac{\partial}{\partial x_i}(u_i P) \tag{3b}$$

with the condition $\phi_i \to 0$ when $u \to \infty$. The following equation for $a_i$ is thus obtained:

$$a_i = \frac{1}{P}\left[\frac{\partial}{\partial u_i}\left(\frac{b_i^2}{2}P\right) + \phi_i\right]. \tag{4}$$

While in the two horizontal directions the PDF is considered to be Gaussian, in the vertical direction the PDF is assumed to be non-Gaussian (to deal with non-uniform turbulent conditions and/or convection). In this latter case, two different approaches can be adopted in order to calculate the Fokker–Planck equation: bi-Gaussian or Gram–Charlier. The bi-Gaussian PDF is given by the linear combination of two Gaussians and the Gram–Charlier PDF is a particular type of expansion that use orthonormal functions in the form of Hermit polynomials.

Comparing the Lagrangian velocity structure functions obtained from Langevin equation with that determined according to Kolmogorov's theory of local isotropy in the inertial subrange, it is possible to determine the diffusion coefficient $b_i = (C_0\varepsilon)^{1/2}$, where $C_0$ is a Kolmogorov constant and $\varepsilon$ is the rate of turbulence kinetic energy dissipation. The product $C_0\varepsilon$ can also be written as a function of the turbulent velocity variance $\sigma_i^2$ and the Lagrangian decorrelation time scale $\tau_{L_i}$ [23,44]:

$$C_0\varepsilon = 2\frac{\sigma_i^2}{\tau_{L_i}}. \tag{5}$$

## 3. Mathematical background—existence and uniqueness

In this section, we present the mathematical background for analyzing the existence and uniqueness of the proposed solutions. Consider the stochastic differential equation

$$\mathrm{d}Z = a(Z(t))\,\mathrm{d}\xi(t) + b(Z(t))\,\mathrm{d}t, \tag{6}$$

where $a : \Re^d \to M(d, r)$ and $b : \Re^d \to \Re^d$ are locally Lipschitz. Then standard results state that pathwise uniqueness of solution holds for (6).

If a global Lipschitz condition holds, it is well known that (6) can be solved using successive approximations (see [24, Section 4.3]). It is well known that under suitable measurability hypotheses,

$$E\left(\exp\left(\frac{1}{2}\int_0^t \|\gamma(\rho, Z)\|\,\mathrm{d}\rho\right)\right) < \infty \quad \forall t > 0$$

and

$$M(t) = \exp\left(\int_0^t \|\gamma(\rho, Z)\|\,\mathrm{d}\xi(s)\right) - \frac{1}{2}\int_0^t \|\gamma(\rho, Z)\|^2\,\mathrm{d}\rho$$

is an adapted martingale. Then, $\tilde{B}(t) = B(t) - \int_0^t \gamma(\rho, Z)\,\mathrm{d}\rho$ is a $d$-dimensional Brownian motion will a new reference probability $\hat{P} = MP$ and

$$\mathrm{d}Z = \alpha d\tilde{B} + (\beta + \gamma)\,\mathrm{d}t \tag{7}$$

(see [24, Theorem 4.2]). That is to say a solution of (7) can be obtained applying the transformation above to the solution of (6) (the so called Girsanov transformation).

If $\alpha < 1$, $\gamma$ and $\beta$ are constants and $Z(0)$ is Gaussian distributed with mean zero and variance $\sigma^2$

$$E(Z(t), Z(s)) = \left(\sigma^2 - \frac{1}{2\gamma}\right)\mathrm{e}^{-\gamma(t+s)} + \frac{1}{2\gamma}\mathrm{e}^{-\gamma(t-s)}, \quad t > s.$$

Then, if $\sigma^2 = 1/2\gamma$, $Z(t)$ is a stationary Gaussian process. In general, if $b = \beta$, the analytic solution of (6) is given by

$$Z(t) = e^{\beta t} Z(0) + \int_0^t e^{-\beta\rho}\, d\beta(\rho). \tag{8}$$

Let us recall that under fairly general circumstances, the stochastic approximations

$$dZ_\delta = a(Z_\delta) \circ d\xi_\delta + b(Z_\delta)\, dt, \tag{9}$$

$Z_\delta = Z$, where $\xi_\delta$ may be taken to be a suitable piecewise-linear approximation to $\xi$, satisfy the estimates

$$\lim_{t\to 0} E(\sup |Z(t, w) - Z_\delta(t, w)|^2) = 0 \tag{10}$$

(see [16, Theorem 7.2]) when $Z$ satisfies (7), $dZ = a(Z)\, d\xi + b(Z)\, dt$, with $\xi$ substituting $\xi_\delta$.

In the next section, we make the choice of some particular PDFs satisfying Eqs. (3a) and (3b), leading to associated Langevin equations. The case of Gaussian PDF leads to the simplest of these equations, namely

$$du_i(t, n) + \alpha_i(n) u_i(t, n) = \beta_i(n) + \gamma(n) u_i^2 + (C_0\varepsilon)^{1/2}\, d\xi(u, t). \tag{11}$$

## 4. Solution for particular PDFs

In pollutant dispersion studies, the turbulence statistics in the PBL can be classified according to time (stationary/non-stationary) and space (homogeneous/inhomogeneous) variations, and according to velocity distribution (Gaussian/non-Gaussian). Using Lagrangian particle models, the turbulence is considered stationary, homogeneous and Gaussian in the two horizontal directions. In the vertical direction, the turbulence is stationary, inhomogeneous and Gaussian or non-Gaussian, according to the stability conditions. In stable and neutral conditions, the velocity distribution can be considered Gaussian. During convective conditions, the velocity distribution is non-Gaussian due to the skewness generated by the organized updrafts and downdrafts motion. In this section, the derivations for the proposed Langevin solution considering Gaussian and non-Gaussian turbulence conditions are presented.

### *4.1. Gaussian PDF [14]*

In Gaussian turbulent flows, a Gaussian PDF is applied in Eq. (4) to obtain the deterministic coefficient of the Langevin (1a):

$$a_i = -\left(\frac{1}{\tau_{L_i}}\right) u_i + \frac{1}{2}\frac{\partial\sigma_i^2}{\partial x_j} + \frac{1}{2\sigma_i^2}\left(\frac{\partial\sigma_i^2}{\partial x_j}\right) u_i^2. \tag{12}$$

Substituting $a_i$ in Langevin (1a) we obtain

$$\frac{du_i}{dt} + \left(\frac{1}{\tau_{L_i}}\right) u_i = \frac{1}{2}\frac{\partial\sigma_i^2}{\partial x_j} + \frac{1}{2\sigma_i^2}\left(\frac{\partial\sigma_i^2}{\partial x_j}\right) u_i^2 + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2} \xi_i(t). \tag{13}$$

Rewriting the Eq. (13) as

$$\frac{du_i}{dt} + \alpha_i u_i = \beta_i + \gamma_i u_i^2 + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2} \xi_i(t), \tag{14}$$

where

$$\alpha_i = \frac{1}{\tau_{L_i}}, \quad \beta_i = \frac{1}{2}\frac{\partial\sigma_i^2}{\partial x_j} \quad \text{and} \quad \gamma_i = \frac{1}{2\sigma_i^2}\left(\frac{\partial\sigma_i^2}{\partial x_j}\right),$$

it is possible to determine $\exp(\alpha_i t)$ as the integrating factor for Eq. (14).

Multiplying the integrating factor by all terms in Eq. (14), one can obtain

$$\frac{\mathrm{d}[\exp(\alpha_i t)u_i]}{\mathrm{d}t} = \beta_i \exp(\alpha_i t) + \gamma_i u_i^2 \exp(\alpha_i t) + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2} \xi_i(t)\exp(\alpha_i t) \tag{15}$$

and, hence,

$$u_i = \exp(-\alpha_i t)\int_{t_0}^{t} [\beta_i \exp(\alpha_i t') + \gamma_i u_i^2 \exp(\alpha_i t') + (2\sigma_i^2/\tau_{L_i})^{1/2}\xi_i(t')\exp(\alpha_i t')]\,\mathrm{d}t'. \tag{16}$$

Picard's Iteration Method is applied to Eq. (16), assuming that the initial guess for the iterative approximation is determined from the stochastic term

$$u_i^0 = \exp(-\alpha_i t)\int_{t_0}^{t} (2\sigma_i^2/\tau_{L_i})^{1/2}\xi_i(t')\exp(\alpha_i t')\,\mathrm{d}t' \tag{17a}$$

and the generic iterative step is written as

$$u_i^{n+1} = \exp(-\alpha_i t)\left[u_i^n + \int_{t_0}^{t} \exp(\alpha_i t')(\beta_i + \gamma_i (u_i^2)^n + (2\sigma_i^2/\tau_{L_i})^{1/2}\xi_i^n(t'))\,\mathrm{d}t'\right], \tag{17b}$$

where the index $n$ represents the number of iteration steps in the Picard Iterative Method.

### 4.2. Bi-Gaussian PDF [14]

If the distribution of vertical turbulent velocity is non-Gaussian, like in a convective PBL, the PDF of the turbulent velocity can be represented by the linear combination of two Gaussian distributions (bi-Gaussian PDF) [4]. Applying a bi-Gaussian PDF in Eq. (4), the expression for the deterministic coefficient of the Langevin equation can be written as

$$a_w = -w\frac{AP_A\sigma_A^2 + BP_B\sigma_B^2}{\sigma_A^2\sigma_B^2}\frac{1}{P}\frac{\sigma_i^2}{\tau_{L_i}} + \left(\frac{Aw_AP_A}{\sigma_A^2} + \frac{Bw_BP_B}{\sigma_B^2}\right)\frac{1}{P}\frac{\sigma_i^2}{\tau_{L_i}} + \frac{\phi}{P}, \tag{18}$$

where $w$ is the vertical turbulent velocity, $A$ and $B$ ($A + B = 1$, $A > 0$, $B > 0$) are, respectively, the proportions of area occupied by updrafts and downdrafts, $w_A$ and $\sigma_A$ are the mean vertical velocity and standard deviation in updrafts, $w_B$ and $\sigma_B$ are the mean vertical velocity and standard deviation in downdrafts and $P_A$ and $P_B$ are Gaussian PDFs and $P$ is the bi-Gaussian PDF of vertical turbulent velocity. The general method for deriving the parameters of the bi-Gaussian PDF, $A$, $B$, $w_A$, $w_B$, $\sigma_A$, $\sigma_B$, consists in equating the zeroth through third moments of $P(z, w)$, $w^m = \int w^m P(z, w)\,\mathrm{d}w$.

Considering the deterministic coefficient (29), the Langevin equation (1a) can be written as

$$\frac{\mathrm{d}w}{\mathrm{d}t} + \frac{AP_A\sigma_A^2 + BP_B\sigma_B^2}{\sigma_A^2\sigma_B^2}\frac{1}{P}\frac{\sigma_i^2}{\tau_{L_i}}w = \left(\frac{Aw_AP_A}{\sigma_A^2} + \frac{Bw_BP_B}{\sigma_B^2}\right)\frac{1}{P}\frac{\sigma_i^2}{\tau_{L_i}} + \frac{\phi}{P} + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2}\xi_w(t), \tag{19}$$

where $\phi$ is obtained applying the bi-Gaussian PDF in Eq. (3b) [29]:

$$\begin{aligned}\phi = & -\frac{1}{2}\left(A\frac{\partial w_A}{\partial z} + w_A\frac{\partial A}{\partial z}\right)\operatorname{erf}\left(\frac{w - w_A}{\sqrt{2}\sigma_A}\right) + w_AP_A\left[A\frac{\partial w_A}{\partial z}\left(\frac{w^2}{\sigma_A^2} + 1\right) + w_A\frac{\partial A}{\partial z}\right] \\ & + \frac{1}{2}\left(B\frac{\partial w_B}{\partial z} + w_B\frac{\partial B}{\partial z}\right)\operatorname{erf}\left(\frac{w + w_B}{\sqrt{2}\sigma_B}\right) + w_BP_B\left[B\frac{\partial w_B}{\partial z}\left(\frac{w^2}{\sigma_B^2} + 1\right) + w_B\frac{\partial B}{\partial z}\right].\end{aligned} \tag{20}$$

Rewriting Eq. (20) as

$$\frac{\mathrm{d}w}{\mathrm{d}t} + \alpha_w w = \beta_w + \gamma_w + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2} \xi_w(t), \tag{21}$$

where

$$\alpha_w = \frac{AP_A\sigma_A^2 + BP_B\sigma_B^2}{\sigma_A^2\sigma_B^2}\frac{1}{P}\frac{\sigma_i^2}{\tau_{L_i}}, \quad \beta_w = \left(\frac{Aw_AP_A}{\sigma_A^2} + \frac{Bw_BP_B}{\sigma_B^2}\right)\frac{1}{P}\frac{\sigma_i^2}{\tau_{L_i}} \quad \text{and} \quad \gamma_w = \frac{\phi}{P},$$

it is possible to determine $\exp(\int_{t_0}^{t} \alpha_w \, \mathrm{d}s)$ as the integrating factor for Eq. (21).

Next, the integrating factor is multiplied by all terms in Eq. (21) to obtain

$$\frac{\mathrm{d}[\exp(\int_{t_0}^{t} \alpha_w \, \mathrm{d}s) w]}{\mathrm{d}t} = \beta_w \exp\left(\int_{t_0}^{t} \alpha_w \, \mathrm{d}s\right) + \gamma_w \exp\left(\int_{t_0}^{t} \alpha_w \, \mathrm{d}s\right) + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2} \xi_w(t) \exp\left(\int_{t_0}^{t} \alpha_w \, \mathrm{d}s\right) \tag{22}$$

and, hence,

$$w = \exp\left(-\int_{t_0}^{t} \alpha_w \, \mathrm{d}s\right)\left[\int_{t_0}^{t} \exp\left(\int_{t_0}^{t'} \alpha_w \, \mathrm{d}s\right)\left(\beta_w + \gamma_w + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2} \xi_w(t')\right) \mathrm{d}t'\right]. \tag{23}$$

The iterative scheme is constructed through Picard's Iteration Method, assuming that the initial value for the wind velocities are random values provided by a non-Gaussian distribution. Therefore, the iterative approach is given by

$$w^{n+1} = \exp\left(-\int_{t_0}^{t} \alpha_w^n \, \mathrm{d}s\right)\left\{w^n + \int_{t_0}^{t} \exp\left(\int_{t_0}^{t'} \alpha_w^n \, \mathrm{d}s\right)\left[\beta_w^n + \gamma_w^n + \left[\left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2}\right]^n \xi_w^n(t')\right] \mathrm{d}t'\right\}. \tag{24}$$

### *4.3. Gram–Charlier PDF [12]*

In Gaussian or non-Gaussian turbulence, a Gram–Charlier PDF can be adopted [1,18]. The Gram–Charlier PDF truncated to the fourth order is given by the following expression [25]:

$$P(r_i) = \frac{\mathrm{e}^{-(r_i^2/2)}}{\sqrt{2\pi}}[1 + C_3H_3(r_i) + C_4H_4(r_i)], \tag{25}$$

where $r_i = u_i/\sigma_i$, $H_3$ and $H_4$ are the Hermite polynomials and $C_3$ and $C_4$ their coefficients. In the case of Gaussian turbulence, Eq. (25) becomes a normal distribution, considering $C_3$ and $C_4$ equal to zero. The third order Gram–Charlier PDF is obtained with $C_4 = 0$.

Applying Eq. (25) in Eq. (4), the deterministic coefficient is given by

$$a_i = \frac{f_i}{h_i}\frac{\sigma_i}{\tau_{L_i}} + \sigma_i\frac{\partial\sigma_i}{\partial x_j}\frac{g_i}{h_i}, \tag{26}$$

where $f_i$, $g_i$ and $h_i$ are expressions written as

$$f_i = -3C_3 - r_i(15C_4 + 1) + 6C_3r_i^2 + 10C_4r_i^3 - C_3r_i^4 - C_4r_i^5, \tag{27a}$$

$$g_i = 1 - C_4 + r_i^2(1 + C_4) - 2C_3r_i^3 - 5C_4r_i^4 + C_3r_i^5 + C_4r_i^6, \tag{27b}$$

$$h_i = 1 + 3C_4 - 3C_3r_i - 6C_4r_i^2 + C_3r_i^3 + C_4r_i^4. \tag{27c}$$

Using the deterministic coefficient given by Eq. (26), we can write the Langevin equation as

$$\frac{\mathrm{d}u_i}{\mathrm{d}t} = \frac{f_i}{h_i}\frac{\sigma_i}{\tau_{L_i}} + \sigma_i\frac{\partial\sigma_i}{\partial x_j}\frac{g_i}{h_i} + \left(\frac{2\sigma_I^2}{\tau_{L_i}}\right)^{1/2} \xi_i. \tag{28}$$

Rewriting Eq. (28) as

$$\frac{\mathrm{d}u_i}{\mathrm{d}t} + \alpha_i u_i = \beta_i + \gamma_i + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2} \xi_i, \tag{29}$$

where

$$\alpha_i = \frac{15C_4 + 1}{h_i \tau_{L_i}}, \quad \beta_i = [f_i + r_i(15C_4 + 1)]\frac{1}{h_i}\frac{\sigma_i}{\tau_{L_i}} \quad \text{and} \quad \gamma_i = \sigma_i \frac{\partial \sigma_i}{\partial x_j}\frac{g_i}{h_i},$$

it is possible to determine $\exp(\int^t \alpha_i \, \mathrm{d}t)$ as the integrating factor for Eq. (29).

Multiplying the integrating factor by all terms in Eq. (29), we can obtain

$$u_i = \exp\left(-\int_{t_0}^{t} \alpha_i \, \mathrm{d}t\right) \left[\int_{t_0}^{t} \exp\left(\int_{t_0}^{t} \alpha_i \, \mathrm{d}t\right) \left(\beta_i + \gamma_i + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2} \xi_i\right) \mathrm{d}t\right]. \tag{30}$$

The Picard Iterative Method is applied to Eq. (30), assuming that the initial value for the turbulent velocity is a random value supplied by a Gaussian distribution. Therefore, the iterative approximation presents the following form:

$$u_i^{n+1} = \exp\left(-\int_{t_0}^{t} \alpha_i^n \, \mathrm{d}t\right) \left\{u_i^n + \int_{t_0}^{t} \exp\left(\int_{t_0}^{t} \alpha_i^n \, \mathrm{d}t\right) \left[\beta_i^n + \gamma_i^n + \left[\left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2}\right]^n \xi_i^n\right] \mathrm{d}t\right\}. \tag{31}$$

## 5. Solution of asymptotic Langevin equation [43]

Under the condition that the Lagrangian decorrelation time scale $\tau_L \to 0$ (or $t/\tau_L \to \infty$), the Langevin model (1a) can be transformed into a stochastic differential equation, which permits to determine the particle positions $x_i$ directly [36]:

$$\mathrm{d}x_i = \left[U_i + \frac{\partial K_i}{\partial x_j}\right] \mathrm{d}t + (2K_i)^{1/2} \xi_i(t) \, \mathrm{d}t, \tag{32}$$

where $K_i$ is the eddy diffusivity. Eq. (32) is called Markov or diffusion equation limit of the Lagevin equation.

The equivalent integral equation of differential Eq. (32) is

$$x_i(t) = \int_{t_0}^{t} \left[U_i + \frac{\partial K_i}{\partial x_j} + (2K_i)^{1/2} \xi_i(t')\right] \mathrm{d}t'. \tag{33}$$

The iterative scheme is generated through Picard's Iteration Method, assuming that the initial value for the positions are random values provided by a Gaussian distribution:

$$x_i^{n+1}(t) = x_i^n(t) + \int_{t_0}^{t} \left[U_i + \frac{\partial K_i}{\partial x_j} + (2K_i)^{1/2} \xi_i(t')\right] \mathrm{d}t'. \tag{34}$$

## 6. Solution for low-wind conditions [13]

When dealing with turbulence and atmospheric dispersion studies, low-wind speeds are generally considered the most critical conditions. The low-wind speed condition is of interest, partly because the simulation of airborne pollutant dispersion in these conditions is rather difficult [2]. During low-wind speed conditions, the diffusion of a pollutant in the PBL is irregular and indefinite. It has been observed that the plume is subject to a great deal of horizontal undulations, which are called plume meandering. The complexity of the boundary layer grows with the weakening of the winds and the degree of atmospheric stability. Thus, it becomes important to study the dispersion in weak wind conditions [40].

The general approach to obtain the model for low-wind dispersion consists in the linearization of the Langevin equation as stochastic differential equation

$$\frac{\mathrm{d}u_i}{\mathrm{d}t} + f(t)u_i = g(t), \tag{35}$$

which has the well known solution determined by the integrating factor $\mathrm{e}^{\int_{t_0}^{t} f(t')\,\mathrm{d}t'}$:

$$u_i = \frac{1}{\mathrm{e}^{\int_{t_0}^{t} f(t')\,\mathrm{d}t'}} \int_{t_0}^{t} g(t')\mathrm{e}^{\int_{t_0}^{t} f(t')\,\mathrm{d}t'}\,\mathrm{d}t'. \tag{36}$$

In order to embody the low wind speed condition in the Langevin equation, it is assumed that the function $f(t)$ is a complex function. Therefore, the exponentials appearing in Eq. (36) can be rewritten like

$$\mathrm{e}^{\int_{t_0}^{t} f(t')\,\mathrm{d}t'} = \mathrm{e}^{\int_{t_0}^{t} p\,\mathrm{d}t' + \int_{t_0}^{t} iq\,\mathrm{d}t'} \tag{37a}$$

or

$$\mathrm{e}^{\int_{t_0}^{t} f(t')\,\mathrm{d}t'} = \mathrm{e}^{pt+\mathrm{i}qt}. \tag{37b}$$

Applying the Euler formula and neglecting the imaginary component, because the wind speed is a real function, Eq. (36) becomes

$$u_i = \mathrm{e}^{-pt}\cos(qt)\int_{t_0}^{t} g(t')\left(\frac{1}{\mathrm{e}^{-pt'}\cos(qt')}\right)\mathrm{d}t'. \tag{38}$$

In Eq. (38), the term $\mathrm{e}^{-pt}\cos(qt)$ is analogous to the Eulerian autocorrelation function suggested in [19] and written in a different way in [3], where $p$ and $q$ are given by

$$p = \frac{1}{(m^2+1)T} \quad \text{and} \quad q = \frac{m}{(m^2+1)T},$$

and $T$ is the time scale for a fully developed turbulence and $m$ is a non-dimensional quantity that controls the meandering oscillation frequency. The autocorrelation function suggested in [19] and in [33] generates a negative lobe for the horizontal wind components attributed to the meander.

Using Lagrangian particle models, the turbulence is considered Gaussian in the horizontal directions ($i = 1, 2$) and, therefore, Eq. (38) can be written as

$$u_i = \mathrm{e}^{-pt}\cos(qt)\int_{t_0}^{t}\left(\frac{1}{\mathrm{e}^{-pt'}\cos(qt')}\right)\left[\beta_i + \gamma_i u_i^2 + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2}\xi_i(t')\right]\mathrm{d}t' \tag{39}$$

where

$$\beta_i = \frac{1}{2}\frac{\partial\sigma_i^2}{\partial x_j} \quad \text{and} \quad \gamma_i = \frac{1}{2\sigma_i^2}\left(\frac{\partial\sigma_i^2}{\partial x_j}\right).$$

The Picard method is applied to Eq. (39), assuming that the initial guess is determined from a Gaussian distribution. The generic iterative step is written as

$$u_i^{n+1} = \mathrm{e}^{-pt}\cos(qt)\left\{u_i^n + \int_{t_0}^{t}\left(\frac{1}{\mathrm{e}^{-pt'}\cos(qt')}\right)\left[\beta_i + \gamma_i(u_i^2)^n + \left(\frac{2\sigma_i^2}{\tau_{L_i}}\right)^{1/2}\xi_i(t')\right]\mathrm{d}t'\right\}. \tag{40}$$

It is important to notice two important situations about Eq. (40). When $m > 0$, the dispersion is under meandering effect, which occur during low-wind conditions. When $m = 0$, the meandering effect is eliminated, that is, the mean wind velocity is more than 1.5–2.0 $\mathrm{m\,s^{-1}}$. In this case, Eq. (40) is written in terms of the exponential form of the autocorrelation function ($e^{-t/\tau_l}$), which Lagrangian particle models usually make use when windy conditions are present. Therefore, the approach (40) is able to simulate the contaminant dispersion in the PBL in both cases, i.e., when the plume evolution is governed by small-scale eddies and exhibits a 'fanning' kind of behavior (typical of windy conditions) and when the plume evolution is governed by large eddies with length and time scale larger than the plume width, causing an undulation (meandering) in its evolution (typical of low-wind conditions).

For applications, the values for the parameters $m$ and $T$ are calculated by the empirical formulation suggested in [35]:

$$m = \frac{8.5}{(1+U)^2}, \quad T = \frac{mT_*}{2\pi(m^2+1)} \quad \text{and} \quad T_* = 200m + 500.$$

## 7. Results and discussion

The performance of the Picard Iterative Method (Eqs. (17), (24), (31), (34) and (40)) have been evaluated against experimental ground-level concentration provided by the Copenhagen [21], Prairie Grass [5], Idaho National Engineering Laboratory (INEL) [38] and Indian Institute of Technology (IIT) [41] diffusion experiments. Furthermore, the results obtained with the proposed approach are compared with the ones predicted by other different models.

For the simulations, the turbulent flow is assumed inhomogeneous only in the vertical ($\partial/\partial x_1 = 0, \partial/\partial x_2 = 0, \partial/\partial x_3 \neq 0$) and the transport is realized by the longitudinal component of the mean wind velocity ($U_1 \neq 0, U_2 = 0, U_3 = 0$). The horizontal domain was determined according to sampler distances and the vertical domain was set equal to the observed PBL height. The time step was maintained constant and it was obtained according to the value of the Lagrangian decorrelation time scale ($\Delta t = \tau_L/c$), where $\tau_L$ must be the smaller value between $\tau_{L_u}, \tau_{L_v}, \tau_{L_w}$ and $c$ is an empirical coefficient set equal to 10. The values of $\sigma_i$, $\tau_{L_i}$ and $K_i$ were parameterized according to the scheme developed in [17]. These parameterizations are based on Taylor's statistical diffusion theory and the observed spectral properties. The third moment of the vertical velocity component is assigned according to Chiba [15] and the fourth moment is calculated from the method suggested in [1]. The concentration field is determined by counting the particles in a cell or imaginary volume in the position $x, y, z$. The integration method used to solve the integrals appearing in Eqs. (17), (24), (31), (34) and (40) was the Romberg technique.

Statistical indices are calculated to compare the simulated and observed concentrations. The statistical indices are the following Hanna [22]:

$$\mathrm{NMSE} = \overline{(C_o - C_p)^2}/\overline{C_o C_p} \quad \text{(normalized mean square error)},$$

$$\mathrm{FB} = (\overline{C_o} - \overline{C_p})/(0.5(\overline{C_o} + \overline{C_p})) \quad \text{(fractional bias)},$$

$$\mathrm{FS} = 2(\sigma_o - \sigma_p)/(\sigma_o - \sigma_p) \quad \text{(fractional standard deviation)},$$

$$R = \overline{(C_o - \overline{C_o})(C_p - \overline{C_p})}/\sigma_o\sigma_p \quad \text{(correlation coefficient)},$$

$$\mathrm{FA2} = \text{fraction of the data for which } 0.5 \leqslant C_p/C_o \leqslant 2 \quad \text{(factor of two)},$$

where $C$ is the analyzed quantity (concentration) and the subscripts "o" and "p" represent the observed and the predicted values, respectively. The overbars in the statistical indices indicate averages. The statistical index FB indicates if the predicted quantity underestimates or overestimates the observed one. The statistical index NMSE represents the quadratic error of the predicted quantity in relation to the observed one. The statistical index FS indicates the measure of the comparison between predicted and observed plume spreading. The statistical index FA2 provides the fraction of data for which $0.5 \leqslant C_o/C_p \leqslant 2$. As nearest zero are the NMSE, FB and FS and as nearest one are the $R$ and FA2, better are the results.

The tracer experiments Copenhagen, Prairie Grass, INEL and IIT are briefly described in the following paragraphs:

Table 1
Meteorological data for the Copenhagen experiment

| Run | $-L$(m) | $h$ (m) | $w_*$ (m s$^{-1}$) | $U$ (10 m) (m s$^{-1}$) | $U$ (60 m) (m s$^{-1}$) | $U$ (115 m) (m s$^{-1}$) |
|---|---|---|---|---|---|---|
| 1 | 37 | 1980 | 1.8 | 2.1 | – | 3.4 |
| 2 | 292 | 1920 | 1.8 | 4.9 | – | 10.6 |
| 3 | 71 | 1120 | 1.3 | 2.3 | – | 5.0 |
| 4 | 133 | 390 | 0.7 | 2.5 | – | 4.6 |
| 5 | 444 | 820 | 0.7 | 3.1 | – | 6.7 |
| 6 | 432 | 1300 | 2.0 | 7.2 | 11.7 | 13.2 |
| 7 | 104 | 1850 | 2.2 | 4.1 | 7.2 | 7.6 |
| 8 | 56 | 810 | 2.2 | 4.2 | 10.6 | 9.4 |
| 9 | 289 | 2090 | 1.9 | 5.1 | 9.0 | 10.5 |

$L$ is the Monin-Obukhov length, $h$ is the PBL height, $w_*$ is the convective velocity scale and $U$ is the mean wind velocity.

Table 2
Meteorological data for the Prairie Grass experiment (unstable case)

| Run | $-L$ (m) | $h$ (m) | $w_*$ (m s$^{-1}$) | $U$ (m s$^{-1}$) |
|---|---|---|---|---|
| 1 | 9 | 260 | 0.84 | 3.2 |
| 5 | 28 | 780 | 1.64 | 7.0 |
| 7 | 10 | 1340 | 2.27 | 5.1 |
| 8 | 18 | 1380 | 1.87 | 5.4 |
| 9 | 31 | 550 | 1.70 | 8.4 |
| 10 | 11 | 950 | 2.01 | 5.4 |
| 15 | 8 | 80 | 0.70 | 3.8 |
| 16 | 5 | 1060 | 2.03 | 3.6 |
| 19 | 28 | 650 | 1.58 | 7.2 |
| 20 | 62 | 710 | 1.92 | 11.3 |
| 25 | 6 | 650 | 1.35 | 3.2 |
| 26 | 32 | 900 | 1.86 | 7.8 |
| 27 | 30 | 1280 | 2.08 | 7.6 |
| 30 | 39 | 1560 | 2.23 | 8.5 |
| 43 | 16 | 600 | 1.66 | 6.1 |
| 44 | 25 | 1450 | 2.20 | 7.2 |
| 49 | 28 | 550 | 1.73 | 8.0 |
| 50 | 26 | 750 | 1.91 | 8.0 |
| 51 | 40 | 1880 | 2.30 | 8.0 |
| 61 | 38 | 450 | 1.65 | 9.3 |

### *7.1. Copenhagen experiment*

The Copenhagen experiment was carried out in the northern part of Copenhagen. The tracer ($SF_6$) was released without buoyancy from a tower at a height of 115 m and collected at the ground-level positions in up to three crosswind arcs of tracer sampling units. The sampling units were positioned 2–6 km from the point of release. A total of nine tracer experiments were performed in stable conditions that are the result of the relative combination between neutral and convective. All available data used to create an input file for the simulations are presented in Table 1. The site was mainly residential with a roughness length of 0.6 m. Wind speeds at 10, 60 and 115 m were used to calculate the coefficient for the exponential wind vertical profile.

### *7.2. Prairie Grass experiment (unstable case)*

The Prairie Grass unstable dispersion experiment was realized in O'Neill, Nebraska, 1956. The pollutant ($SO_2$) was emitted without buoyancy at a height of 0.5 m and it was measured by samplers at a height of 1.5 m in five downwind distances (50, 100, 200, 400, 800 m). The Prairie Grass site was flat with a roughness length of 0.6 cm. The results for

Table 3
Meteorological data for the Prairie Grass experiment (stable case)

| Run | $L$ (m) | $h$ (m) | $u_*$ (m s$^{-1}$) | $U$ (m s$^{-1}$) |
|---|---|---|---|---|
| 13 | 3.4 | 23 | 0.09 | 3.9 |
| 14 | 1.6 | 12 | 0.05 | 3.7 |
| 17 | 48 | 131 | 0.21 | 3.8 |
| 18 | 25 | 92 | 0.2 | 4 |
| 21 | 172 | 333 | 0.38 | 6.4 |
| 22 | 204 | 400 | 0.46 | 7.7 |
| 23 | 193 | 358 | 0.39 | 6.5 |
| 24 | 248 | 400 | 0.38 | 6.3 |
| 28 | 24 | 81 | 0.16 | 3.2 |
| 29 | 36 | 119 | 0.23 | 4.3 |
| 32 | 8.3 | 43 | 0.13 | 3.6 |
| 35 | 53 | 147 | 0.24 | 4.3 |
| 36 | 7.8 | 36 | 0.1 | 2.8 |
| 37 | 95 | 216 | 0.29 | 5 |
| 38 | 99 | 217 | 0.28 | 4.8 |
| 39 | 9.8 | 48 | 0.14 | 3.6 |
| 40 | 8 | 39 | 0.11 | 3.1 |
| 41 | 35 | 117 | 0.23 | 4.4 |
| 42 | 120 | 275 | 0.37 | 6.3 |
| 46 | 114 | 257 | 0.34 | 5.8 |
| 53 | 10 | 54 | 0.17 | 4.3 |
| 54 | 40 | 128 | 0.24 | 4.5 |
| 55 | 124 | 279 | 0.37 | 6.3 |
| 56 | 76 | 194 | 0.29 | 5.1 |
| 58 | 6.4 | 35 | 0.11 | 3.4 |
| 59 | 11 | 51 | 0.14 | 3.4 |
| 60 | 58 | 166 | 0.28 | 5 |

$u_*$ is the friction velocity.

twenty convective experiments are presented. The all available data used to create a input file for the simulations are presented in Table 2. Wind speed profile has been parameterized following the similarity theory of Monin-Obukhov and OML model [6].

### 7.3. Prairie Grass experiment (stable case)

The Prairie Grass stable dispersion experiment was carried out in O'Neill, Nebraska, 1956. The tracer ($SO_2$) was released without buoyancy at a height of $\sim$ 0.5 m, and colleted at a height of 1.5 m at three downwind distances (50, 200 and 800 m). The Prairie Grass site was quite flat and much smooth with a roughness length of 0.6 cm. The micrometeorological parameters recorded during the dispersion experiments are summarized in Table 3. Exponential wind vertical profiles were calculated from the wind speed measured during the experiment.

### 7.4. INEL experiment (stable low-wind case)

The Idaho National Engineering Laboratory (INEL) experiment was conducted during stable low-wind conditions. Because of wind direction variability, a full 360° sampling grid was implemented. Arcs were laid out at radii of 100, 200 and 400 m from the emission point. Samplers were placed at intervals of 6° on each arc for a total of 180 sampling positions. The receptor height was 0.76 m. The tracer $SF_6$ was released at a height of 1.5 m. Table 4 shows the meteorological data utilized for the validation of the proposed models. Wind speeds at levels 2, 4, 8, 16, 32 and 61 m were used to calculate the coefficient for the exponential wind vertical profile. Following Brusasca et al. [8] and Sharan and Yadav [42], the roughness length utilized was $z_0 = 0.005$ m.

Table 4
Meteorological data for the INEL experiment

| Run | $L$ (m) | $h$ (m) | $u_*$ (m s$^{-1}$) | $U$ (2 m) (m s$^{-1}$) | $U$ (4 m) (m s$^{-1}$) | $U$ (8 m) (m s$^{-1}$) | $U$ (16 m) (m s$^{-1}$) | $U$ (32 m) (m s$^{-1}$) | $U$ (61 m) (m s$^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|
| 4 | 2.40 | 13.40 | 0.047 | 0.7 | 1.2 | – | 1.5 | 0.9 | 2.1 |
| 5 | 3.14 | 16.38 | 0.053 | 0.8 | 0.9 | 1.2 | 2.2 | 3.0 | 2.1 |
| 7 | 1.77 | 10.64 | 0.040 | 0.6 | 0.9 | 0.4 | 0.5 | 0.9 | 2.4 |
| 8 | 1.22 | 8.09 | 0.033 | 0.5 | 0.8 | 0.6 | 1.2 | 1.6 | 2.7 |
| 9 | 1.22 | 8.09 | 0.033 | 0.5 | 0.8 | 0.9 | 1.6 | 2.2 | 2.7 |
| 10 | 5.93 | 26.40 | 0.073 | 1.1 | 1.7 | 2.1 | 3.2 | 4.7 | 3.1 |
| 11 | 9.60 | 37.91 | 0.093 | 1.4 | 1.9 | 2.3 | 2.9 | – | 3.6 |
| 12 | 2.40 | 13.40 | 0.047 | 0.7 | 1.1 | 1.1 | 1.6 | 1.6 | 1.9 |
| 13 | 4.90 | 22.88 | 0.067 | 1.0 | 1.6 | 2.0 | 3.0 | 4.0 | 6.0 |
| 14 | 4.90 | 22.88 | 0.067 | 1.0 | 1.5 | 2.0 | 3.5 | 5.1 | 7.1 |

Table 5
Meteorological data for the IIT experiment

| Run | $-L$ (m) | $h$ (m) | $w_*$ (m s$^{-1}$) | $U$ (m s$^{-1}$) |
|---|---|---|---|---|
| 1 | 17.5 | 1570 | 2.37 | 1.36 |
| 2 | 2.5 | 1240 | 2.26 | 0.74 |
| 6 | 20 | 1070 | 2.04 | 1.40 |
| 7 | 22 | 1240 | 2.28 | 1.54 |
| 8 | 32 | 943 | 1.09 | 0.89 |
| 11 | 13 | 1070 | 1.83 | 1.07 |
| 12 | 24 | 1325 | 2.32 | 1.55 |
| 13 | 2 | 1070 | 1.72 | 1.08 |

Table 6
Statistical evaluation of the proposed model and comparison with other dispersion models

| Experiment | Model | NMSE | FB | FS | R | FA2 |
|---|---|---|---|---|---|---|
| Copenhagen | Eq. (17) | 0.05 | −0.11 | −0.11 | 0.93 | 1.00 |
| | Eq. (24) | 0.04 | −0.11 | 0.01 | 0.94 | 0.96 |
| | Eq. (31) | 0.03 | 0.01 | 0.03 | 0.93 | 1.00 |
| | Eq. (34) | 0.03 | 0.05 | 0.02 | 0.94 | 1.00 |
| | Ito—Gaussian [9] | 0.06 | 0.09 | 0.27 | 0.91 | 1.00 |
| | Ito—bi-Gaussian [10] | 0.07 | −0.14 | 0.01 | 0.92 | 1.00 |
| | Ito–Gram–Charlier [11] | 0.08 | −0.02 | −0.06 | 0.82 | 0.96 |
| | Eulerian [46] | 0.07 | 0.06 | 0.23 | 0.90 | 1.00 |
| | Gaussian [16] | 0.08 | 0.10 | 0.31 | 0.87 | 1.00 |
| Prairie Grass—unstable | Eq. (31) | 0.10 | 0.05 | −0.06 | 0.96 | 0.72 |
| | Ito—Gram–Charlier [11] | 0.08 | 0.008 | 0.07 | 0.96 | 0.78 |
| | Eulerian [31] | 0.09 | −0.21 | 0.14 | 0.98 | 0.61 |
| | Gaussian [9] | 0.27 | −0.22 | −0.36 | 0.95 | 0.81 |
| Prairie Grass—stable | Eq. (17) | 0.39 | −0.057 | −0.18 | 0.81 | 0.95 |
| | Ito—Gaussian [30] | 0.66 | −0.15 | −0.39 | 0.77 | 0.94 |
| | Eulerian [30] | 0.49 | −0.22 | −0.26 | 0.77 | 0.82 |
| INEL | Eq. (40) | 0.11 | 0.02 | −0.18 | 0.93 | 0.83 |
| | Sagendorf and Dickson [38] | 0.60 | 0.06 | – | 0.42 | 0.80 |
| | Sharan and Yadav [42] | 0.53 | −0.02 | – | 0.55 | 0.60 |
| | Oettl et al. [34] | 0.21 | −0.13 | – | 0.86 | 0.87 |
| | Moreira et al. [32] | 0.25 | 0.02 | 0.08 | 0.79 | 0.79 |
| IIT | Eq. (40) | 0.08 | 0.004 | −0.13 | 0.93 | 0.88 |
| | Aria [3] | 13.86 | 1.68 | 1.59 | 0.77 | – |
| | Sharan et al. [41] | 7.11 | 1.49 | 1.32 | 0.76 | – |
| | Moreira et al. [32] | 0.35 | −0.01 | −0.033 | 0.76 | 0.81 |

Table 7
Computational time comparison between the proposed and the classical solution of the Langevin equation (integrated according to the Ito calculus) as a function of the number of released particles in each time step

| Computational time (s) | | |
|---|---|---|
| Number of particles | Gaussian PDF Eq. (17) | Ito Gaussian |
| (a) | | |
| 30 | 56 | 66 |
| 40 | 76 | 87 |
| 50 | 95 | 109 |
| Number of particles | bi-Gaussian PDF Eq. (24) | Ito bi-Gaussian |
| (b) | | |
| 30 | 115 | 135 |
| 40 | 153 | 177 |
| 50 | 193 | 222 |
| Number of particles | Gram–Charlier PDF Eq. (31) | Ito Gram–Charlier |
| (c) | | |
| 30 | 114 | 134 |
| 40 | 153 | 176 |
| 50 | 192 | 223 |
| Number of particles | Asymptotic solution Eq. (34) | Ito asymptotic |
| (d) | | |
| 30 | 34 | 56 |
| 40 | 45 | 76 |
| 50 | 56 | 95 |

(a) Gaussian PDF, (b) bi-Gaussian PDF, (c) Gram–Charlier PDF and (d) asymptotic Langevin equation.

### *7.5. IIT experiment (unstable low-wind case)*

The Indian Institute of Technology (IIT) experiment was conducted during convective low-wind conditions. The pollutant ($SF_6$) was released without buoyancy at a height of 1 m and the concentrations were observed at a height of 0.5 m. The release rate of $SF_6$ tracer varied between 30 to 50 ml min$^{-1}$. Wind and temperature measurements were obtained at four levels (2, 4, 15 and 30 m) from 30 m micrometeorological tower. In all the cases, the wind speed was less than 2 m s$^{-1}$. The samplers were located on arcs of 50 and 100 m radii. Table 5 shows the meteorological data utilized for the model validation.

Model results are shown in Tables 6 and 7. Table 6 shows the result of the statistical analysis made with observed and predicted values of ground-level concentration following Hanna's (1989) [22] statistical indices. Table 7 presents the computational time comparison between the proposed and the classical solutions of the Langevin equation (integrated according to the Ito calculus). Analyzing the results showed in Table 6 it is possible to observe that new approach, independent of the PDF type, presents good agreements with the observational data. It is also possible to verify that results generated by the new solution are comparable or even better than ones obtained by the Langevin equation integrated according to the Ito calculus and other models. The statistical analysis presents NMSE, FB and FS values relatively near to zero and $R$ and FA2 relatively near to 1.

Specifically, we promptly realized that the asymptotic solution (random displacement model) generates accurate results with a lesser computational effort when compared with other models. Therefore, we observe additional advantages of this method over either of the approaches. The explanation for these improvements are based on the argument that the random displacement model solves only one equation (displacement equation); meanwhile the proposed iterative solution and Ito model solve two equations (velocity and displacement equations). As a consequence, bearing in mind the semi-analytical feature of these solutions, we may affirm that the round-off error effect is reduced in the approach because the number of computational operations is reduced. It is important to observe that it is possible to use time steps larger than $\Delta t = \tau_L / c$ in random displacement models, if numerical error due to the inhomogeneity of the turbulent or mean properties of the flow is not introduced [37]. In this sense, the computational time in the asymptotic model could be reduced in relation to the values presented in Table 7.

The results obtained by the proposed solution for low-wind conditions agree very well with the experimental data, indicating the model represents the dispersion process correctly in low-wind speed conditions. It is also possible to verify that results obtained by the new solution are better than ones obtained by the other models. The analytical feature of the proposed technique and the natural inclusion of the Eulerian autocorrelation function suggested in [19] make the model more accurate than other models. These results show that the model can be also used in low-wind speed events happening under unstable conditions.

Under the mathematical point of view, it is relevant to consider two aspects about the proposed iterative method. First, the well-known Picard Iteration Method is considered here to solve integral equation, assuming that the issues regarding existence and uniqueness are fulfilled. Second, it is relevant to point out that the major advantage relies on the fact that it solves iteratively the Langevin equation and the velocity is evaluated analytically in each iteration.

Bearing in mind the statistical evaluation and the aspects mentioned above, the new technique can be considered an alternative approach to solve the Langevin equation. Picard's Iteration Method employed in the proposed approach is a well-known procedure to solve integral equations but to our knowledge it had not been applied in the solution of the Langevin equation. The results obtained are encouraging, showing that it may be incorporated in a modelling system for air-quality estimates.

## References

[1] D. Anfossi, E. Ferrero, D. Sacchetti, S. Trini Castelli, Comparison among empirical probability density functions of the vertical velocity in the surface layer based on higher order correlations, Boundary-Layer Meteorology 82 (1997) 193–218.

[2] D. Anfossi, D. Oettl, G.A. Degrazia, A. Goulart, An analysis of sonic anemomenter observations in low wind speed conditions, Boundary-Layer Meteorology 1 (114) (2005) 179–203.

[3] P. Arya, Modeling and parameterization of near-source diffusion in weak winds, J. Appl. Meteorology 34 (1995) 1112–1122.

[4] J.H. Baerentsen, R. Berkowicz, Monte Carlo simulation of plume dispersion in the convective boundary layer, Atmospheric Environ. 18 (1984) 701–712.

[5] M.L. Barad, Project Prairie Grass: a field program in diffusion, Geophys. Res. Paper No. 59 (II) TR-58-235 (II), Air Force Cambridge Research Centre, USA, 1958.

[6] R.R. Berkowicz, H.R. Olesen, U. Torp, The Danish Gaussian air pollution model (OML): description, test and sensitivity analysis in view of regulatory applications, in: C. De Wispeleare, F.A. Schiermeirier, N.V. Gillani (Eds.), Air Pollution Modeling and Its Application, Plenum Publishing Corporation, 1986, pp. 453–480.

[7] W. Boyce, R. Di Prima, Elementary Differential Equations and Boundary Value Problems, Wiley, 2001, 749pp.

[8] G. Brusasca, G. Tinarelli, D. Anfossi, Particle model simulation of diffusion in low wind speed stable conditions, Atmospheric Environ. 26a (1992) 707–723.

[9] J.C. Carvalho, Aplicação do modelo de partículas Lagrangeano LAMBDA em codições de turbulência Gaussiana, 2003, personal communication.

[10] J.C. Carvalho, G.A. Degrazia, D. Anfossi, Simulation of compenhagen tracer diffusion experiment by means of a Lagrangian particle model, Hybrid Methods in Engineering 01 (04) (1999) 309–327.

[11] J.C. Carvalho, G.A. Degrazia, D. Anfossi, C.R.J. Campos, D. Roberti, A. Kerr, Lagrangian stochastic dispersion modelling for the simulation of the release of contaminants from tall and low sources, Meteorologische Z. 11 (2) (2002) 89–97.

[12] J.C. Carvalho, E.R. Nichimura, M.T. Vilhena, D.M. Moreira, G.A. Degrazia, An iterative langevin solution for contaminant dispersion simulation using the Gram–Charlier PDF, Environ. Model Softw. 20 (3) (2005) 285–289.

[13] J.C. Carvalho, M.T. Vilhena, Pollutant dispersion simulation for low wind speed condition by the ILS method, Atmospheric Environ. 39 (2005) 6282–6288.

[14] J.C. Carvalho, M.T. Vilhena, D.M. Moreira, An alternative numerical approach to solve the Langevin equation applied to air pollution dispersion, Water Air and Soil Pollution 163 (1–4) (2005) 103–118.

[15] O. Chiba, Stability dependence of the vertical wind velocity skewness in the atmospheric surface layer, J. Meteorological Soc. Japan 56 (1978) 140–142.

[16] G.A. Degrazia, Modelling dispersion from elevated sources in a planetary boundary layer dominated by moderate convection, Il Nuovo Cimento 21C (3) (1998) 345–353.

[17] G.A. Degrazia, D. Anfossi, J.C. Carvalho, C. Mangia, T. Tirabassi, H.F. Campos Velho, Turbulence parameterization for PBL dispersion models in all stability conditions, Atmospheric Environ. 34 (2000) 3575–3583.

[18] E. Ferrero, D. Anfossi, Comparison of PDFs, closures schemes and turbulence parameterizations in Lagrangian stochastic models, Internat. J. Environ. Pollution 9 (1988) 384–410.

[19] F.N. Frenkiel, Turbulent diffusion: Mean concentration distribution in a flow field of homogeneous turbulence, Adv. Appl. Mech. 3 (1953) 61–107.

[20] C.W. Gardiner, Handbook of Stochastic Methods for Physics, Chemistry and the Natural Sciences, Springer, Berlin, 1985.
[21] S.E. Gryning, E. Lyck, Atmospheric dispersion from elevated source in un urban area: comparison between tracer experiments and model calculations, J. Climate Appl. Meteorology 23 (1984) 651–654.
[22] S.R. Hanna, Confidence limit for air quality models as estimated by bootstrap and jacknife resampling methods, Atmospheric Environ. 23 (1989) 1385–1395.
[23] J.O. Hinze, Turbulence, McGraw-Hill, New York, 1975.
[24] N. Ikeda, S. Watanabe, Stochastic Differential Equations and Diffusion Process, North-Holland, Amsterdam, 1981.
[25] M. Kendall, A. Stuart, The Advanced Theory of Statistics, MacMillan, New York, 1977.
[26] B.J. Legg, M.R. Raupach, Markov chain simulation of particle dispersion in inhomogeneous flows: the mean drift velocity induced by a gradient in Eulerian velocity variance, Boundary-Layer Meteorology 24 (1982) 3–13.
[27] C.C. Lin, W.H. Reid, Turbulent flow, theroretical aspects, Handbuch Physik, der vol. VIII/2, 1963, pp. 438–523.
[28] A.K. Luhar, R.E. Britter, A random walk model for dispersion in inhomogeneous turbulence in a convective boundary layer, Atmospheric Environ. 23 (1989) 1191–1924.
[29] A.K. Luhar, M.F. Hibberd, P.J. Hurley, Comparison of closure schemes used to specify the velocity PDF in Lagrangian stochastic dispersion models for convective conditions, Atmospheric Environ. 30 (1996) 1407–1418.
[30] D.M. Moreira, J.C. Carvalho, A. Goulart, A comparison of two turbulent dispersion parameterisation in the stable boundary layer by using Lagrangian and Eulerian model, Revista Brasileira de Geofísica 20 (2) (2002) 103–112.
[31] D.M. Moreira, G.A. Degrazia, M.T. Vilhena, Dispersion from low sources in a convective boundary layer: an analytical model, Il Nuovo Cimento 22C (1999) 685–691.
[32] D.M. Moreira, T. Tirabassi, J.C. Carvalho, Plume dispersion simulation in low wind conditions in the stable and convective boundary layers, Atmospheric Environ. 39 (2005) 3643–3650.
[33] R.J. Murgatroyd, Estimations from geostrophic trajectories of horizontal diffusivity in the mid-latitude troposphere and lower stratosphere, Quart. J. Roy. Meteorological Soc. 95 (1969) 40–62.
[34] D. Oettl, R.A. Almbauer, P.J. Sturm, A new method to estimate diffusion in stable, low-wind conditions, J. Appl. Meteorology 40 (2001) 259–268.
[35] D. Oettl, D. Anfossi, Private Communication, 2005.
[36] H.C. Rodean, Stochastic Lagrangian models of turbulent diffusion, American Meteorological Society, Boston, 1996.
[37] H.C. Rodean, R. Lange, J.S. Nasstrom, Comparison of Two Stochastic Models of Scalar Diffusion in Turbulent Flow, Lawrence Livermore National Laboratory UCRL-JC-110377, 1992.
[38] J.F. Sagendorf and C.R. Dickson, Diffusion under low wind-speed, inversion conditions, U.S. National Oceanic and Atmospheric Administration Technical Memorandum ERL ARL-52, 1974.
[39] K. Shankar Rao, Lagrangian stochastic modeling of dispersion in the stable boundary layer, Boundary-Layer Meteorology 90 3 (1999) 541–549.
[40] M. Sharan, S.G. Gopalakrishnan, Mathematical modeling of diffusion and transport of pollutants in the atmospheric boundary layer, PAGEOPH 160 (2003) 357–394.
[41] M. Sharan, M.P. Singh, A.K. Yadav, A mathematical model for the atmospheric dispersion in low winds with eddy diffusivities as linear functions of downwind distance, Atmospheric Environ. 30 (7) (1996) 1137–1145.
[42] M. Sharan, A.K. Yadav, Simulation of experiments under light wind, stable conditions by a variable K-theory model, Atmospheric Environ. 32 (1998) 3481–3492.
[43] C.R.P. Szinvelski, M.T.M.B. Vilhena, J.C. Carvalho, G.A. Degrazia, Semi-analytical solution of the asymptotic langevin equation by the picard iterative method, Environ. Model Softw. 21 (3) (2006) 406–410.
[44] H. Tennekes, Similarity relation, scaling laws and spectral dynamics, in: F.T.M. Nieuwstadt, H. Van Dop (Eds.), Atmospheric Turbulence and Air Pollution Modeling, Reidel, Dordrecht, 1982, pp. 37–68.
[45] D.J. Thomson, Criteria for the selection of stochastic models of particle trajectories in turbulent flows, J. Fluid Mech. 180 (1987) 529–556.
[46] M.T. Vilhena, U. Rizza, G.A. Degrazia, C. Mangia, D.M. Moreira, T. Tirabassi, An analytical air pollution model: development and evaluation, Contrib. Atmospheric Phys. 71 (3) (1998) 315–320.
[47] J.C. Weil, P.P. Sullivan, C.-H. Moeng, The use of large-eddy simulation in Lagrangian particle dispersion models, J. Atmospheric Sci. 61 (2004) 2877–2887.